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Neutron Fission of $^{235,237,239}U$ and $^{241,243}Pu$: Cross Sections, Integral Cross Sections and Cross Sections on Excited States

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Neutron Fission of ^{235,237,239}U and ^{241,243}Pu: Cross Sections, Integral Cross Sections and Cross Sections on Excited States

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I. INTRODUCTION

In a recent paper [1, 2] submitted to Phys. Rev. C we have presented estimates for (n, f) cross sections on a series of Thorium, Uranium and Plutonium isotopes over the range $E_n = 0.1 - 2.5$ MeV. The (n, f) cross sections for many of these isotopes are difficult or impossible to measure in the laboratory. The cross sections were obtained from previous (t, pf) reaction data invoking a model which takes into account the differences between (t, pf) and (n, f) reaction processes, and which includes improved estimates for the neutron compound formation process.

The purpose of this note is: (1) to compare the estimated cross sections to current data files in both ENDF [3, 4] and ENDL [5] databases; (2) to estimate ratios of cross sections relative to ^{235}U integrated over the "tamped flattop" critical assembly spectrum that was used in our earlier ^{237}U report [6] and; (3) to show the effect on the integral cross sections when the neutron capturing state is an excited rotational state or an isomer. The isomer and excited state results are shown for ^{235}U and ^{237}U.

II. COMPARISONS TO ENDF AND ENDL

In [1, 2] we showed only comparisons to ENDF/B-VI and did not include ENDL since that database is not broadly available. ENDL is, however, an important database for LLNL programs. Additionally, since the writing of [1, 2], a new ENDF/B-VII evaluation for ^{237}U and ^{239}U has become available [4], which is somewhat different from ENDF/B-VI. In the following we will also make comparisons with that evaluation.

Fig. 1 shows estimated cross sections for ^{235,237,239}U compared to the ENDL, ENDF/B-VI, and ENDF/B-VII evaluations. The ^{235}U data agree quite well with evaluations and with experimental data for the region above about $E_n = 0.5$ MeV. In the region $0.1 < E_n < 0.5$ MeV the estimates are high by up to 20% which may be due to the estimated neutron compound cross sections or to some details of the low-lying fission transitions states [1, 2].

For ^{237}U our results are generally flatter and lower than any of the three evaluations. However, as discussed in our ^{237}U Report [6], our results are in good agreement with a previous critical assembly measurement [7] and with the McNally measurement [8], if it is assumed that the ^{237}Np contamination in their sample is higher than their original estimate. This subject was discussed in more detail previously [6]. The cross-section comparison is included here for completeness.

For ^{239}U our results are flatter than either the ENDL or ENDF/B-VII evaluations and they lie in a region between the two evaluations. Our results are the only "experimental" data likely to ever be available for this nucleus because of its short, 23-min, half-life. Future work on the model might affect results below $E_n = 0.5$ MeV as discussed for ^{235}U, but this is expected to be at most a 20% effect.

Fig. 2 shows estimated cross sections for ^{241,243}Pu. For ^{241}Pu our results are systematically lower than ENDL, ENDF/B-VI and the previous experimental data by about 15%, but show a similar shape, except for possibly rising too fast below $E_n = 0.5$ MeV. This is consistent with the ^{235}U results. For the five cases presented in our Phys. Rev. C paper [1, 2] where good experimental data exist, this was the case with the maximum deviation. This comparison led us to the conclusion that the estimated $\pm 10\%$ systematic errors for absolute fission probabilities in the (t, pf) experiments is realistic. The same $\pm 10\%$ systematic uncertainty applies to the deduced (n, f) cross sections.

For ^{243}Pu our experimental results are very different from the ENDL and ENDF/B-VI databases which appear to have a common origin. The evaluations have an unphysical shape and it is not clear how they were obtained. Because of the 5-hr half-life for ^{243}Pu, it is unlikely that the results in [1, 2] can be improved upon, except for possible less-than-20% effects in the region below $E_n = 0.5$ MeV, as discussed above.

III. FISSION FROM CAPTURE ON EXCITED STATES

In some environments, fission can involve neutron capture on a short-lived excited state rather than on the ground state. The most important example of this situation is the $T_{1/2} \approx 26$-minute, $J^{\pi} = 1/2^{+}$ isomer in ^{235}U. In our earlier paper [9] we have studied this case and shown that the fission cross section for the isomer is reduced by up to 30% at the lowest energies. Above 1 MeV the cross sections for the ground state ($7/2^{-}$) and isomer become virtually identical. The low energy suppression for fission from the isomer is a nuclear structure effect related to the hindrance of fission near the barrier for even-even nuclei with spins 1^{+} or 0^{-}.

In a manner similar to the isomer calculation it is also possible to estimate fission cross sections for the low-lying rotational states which might be important in some circumstances. Fig. 3 shows estimated (n, f) cross sections for the band head and the first two rotational excited states built on ^{235}U^{gs}, ^{235}U^{m}, and ^{237}U^{gs} states. Note that ground state of ^{237}U has the same $1/2^{+}$ spin/parity as the isomer in ^{235}U and so it is expected that its fission will be hindered at low energies. At low energies the cross sections rise going from the $J^{\pi} = 1/2^{+}$ to the $3/2^{+}$ and then $5/2^{+}$ rotational states. This is because the relative population of the hindered 1^{+} and 0^{-} compound states is decreased. For ^{235}U^{gs} ($J^{\pi} = 7/2^{-}$) we see very little variation of the estimated (n, f) cross-section as we go up the rotational band, $J^{\pi} = 7/2^{-}$, $9/2^{-}$, $11/2^{-}$.

The relative importance of these small effects is better illustrated by folding the cross sections with the flux from a critical assembly, which we do in the next section.

IV. INTEGRAL CROSS SECTIONS

In our ^{237}U report [6] we compared the ratio of the ^{237}U to ^{235}U cross sections integrated over the "tamped flattop" spectrum from previous LANL critical assembly experiments [7]. The soft tamped spectrum was used because of the energy range of our data is limited to $0.1 < E_n < 2.5$ MeV.

Figs. 4 and 5 show the integral of our cross sections along with ENDL and ENDF/B evaluations folded with the "tamped flattop" critical assembly flux used by Barr [7] in his measurement for ^{237}U. This integral is taken from 0.1 MeV to 2 MeV and is shown for all of the isotopes. Fig. 6 shows the same procedure applied to the ^{235}U and ^{237}U excited states.

The integral results show different behaviors for the two cases where there is good experimental data . In ^{235}U the overestimate for our results below 0.5 MeV, which contains much of the tamped flattop flux, leads to an integral that is higher than corresponding the value for ENDL or ENDF/B-VI. In ^{241}Pu the underestimate of the (n, f) cross section at energies above $E_n = 1$ MeV leads to a corresponding underestimate of the integral.

In order to quantify these results further we have taken the ratio to ^{235}U for the tamped flattop integral estimates. This ratio, R_{235}, is the normally-measured quantity in the critical assembly studies. The results for R_{235} for the isotopes studied here are shown in Table I. In these ratios the ^{235}U cross sections are taken from our data set for use with our data, and from ENDL or ENDF, for use with data from those evaluations.

For ^{237}U our value of 0.39 is in very good agreement with the measured value 0.391 [7]. For comparison, the previous estimate of R_{235} for ^{237}U [6] used the ENDF/B-VI cross sections for the ^{235}U denominator and produced a value of 0.43. A recent re-examination of this analysis by Wilhelmy [10] concluded that our values may be overestimated by the order of 10% from experimental data because of the restricted energy range (0.1 − 2.0 MeV) that was available from our data. In summary, we feel that the current values of R_{235} for ^{237}U and the other isotopes should be accurate to about 20%, which is the estimated systematic uncertainty in the absolute fission probabilities that underlie these results and in our model for $E_n \leq 0.5$ MeV [1, 2].

Comparing R_{235} to the current evaluated data files for ^{237}U we see in Fig. 1b) and Table I that there are serious discrepancies. The R_{235} value for ENDL is about 75% higher than ours or the Barr results [7]. Also, in Fig. 1b), ENDL shows a shape for the energy dependence that has a physically unrealistic structure. For ENDF/B-VII the value of R_{235} is closer to ours (10% higher) but again the shape of the cross section seems physically unreasonable. Some of the shape problems may have come from the evaluators trying to reproduce the high energy part of the reported McNally data [8] which we believe is contaminated by contributions from a ^{237}Np background.

For ^{239}U our results are the only experimental data available. The ENDL and ENDF/B-VI values are of unknown origin and their R_{235} values overshoot and undershoot our results by a factor of 2, respectively.

For ^{241}Pu our result is 20% below the evaluated data sets. This is consistent with the comparison of the (n, f) cross section averaged over the region 0.1 to 2.5 MeV where we are about 15% low [1, 2]. Of the five cases we studied where our results overlap large experimental data sets, ^{241}Pu is the extreme deviation [1, 2].

For ^{243}Pu our results again are the only experimental data available. The evaluations in ENDL and ENDF/B-VI appear identical, are of unknown origin and clearly have an unreasonable, unphysical shape below $E_n = 1.5$ MeV.

For the excited states, the R_{235} values show some interesting systematics. The only significant effect is the decrease by about 15% for fission through the $^{235}U^m$ isomer relative to fission through the ground state. Similarly the ^{237}U ground state probably has the same hindrance of fission relative to what it would have been if the ground state spin were different.

Within the $J^\pi = 1/2^+$ bands there may be two small effects of opposite sign at work. First as the spin increases from $J^\pi = 1/2$ to $3/2$ and then $5/2$ the relative population of 1^+ state in the compound nucleus should decrease allowing the fission rate to increase. A second opposite effect is due to the increased spacing of the rotational levels at the fission saddle points relative to the ground-state rotational band. This has the effect of slightly increasing the fission barrier as the angular momentum increases and leads toward a decrease in the fission rate. From the results in Table I it appears that these two effects roughly cancel for the $J^\pi = 1/2^+$ bands and the level spacing effect alone has only a small effect for the $J^\pi = 7/2^-$ band. In summary, our results indicate that there is no significant change in fission rate depending on whether the capturing nucleus is in its ground state or a higher rotational level in the same band for a given nucleus.

The main parameter in determining the magnitude of the fission cross section relative to the total reaction cross section is the difference in energy between the neutron binding energy and the fission barrier height. Fig. 7 shows a plot of the R_{235} factors as a function of the neutron binding energy minus the estimated height of the first peak of the fission barrier [11], $B_n - E_A$. In all of these cases barrier A is estimated to be the highest one although in the U isotopes the two fission barriers have relatively equal heights for the isotopes of interest here. This plot shows a correlation of R_{235} with $B_n - E_A$, which is expected from simple statistical models of the competition between fission and neutron emission [12].

REFERENCES

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[1] W. Younes and H. C. Britt (2003), submitted to Phys. Rev. C.

[2] W. Younes and H. C. Britt, *Simulated neutron-induced fission cross sections for various Pu, U and Th isotopes*, Tech. Rep. UCRL-JC-153292, LLNL (2003).

[3] Cross Section Evaluation Working Group, *ENDF/B-VI Summary Documentation,* Report **BNL-NCS-17541 (ENDF-201)** (1991), edited by P. F. Rose, National Nuclear Dat Center, Brookhave Laboratory, Upton, NY, USA.

[4] P. G. Young and M. B. Chadwick, *$^{237,239,241}U + n$ Evaluations*, Tech. Rep. Los Alamos Memo, T-16:NW-2/10-00, LANL (2003).

[5] *Evaluated neutron data library*, www-ndg.llnl.gov.

[6] W. Younes, H. C. Britt, and J. B. Wilhelmy, *The $^{237}U(n, f)$ cross section*, Tech. Rep. UCRL-ID-152621, LLNL (2003).

[7] D. W. Barr, private communication as quoted in ref. [8].

[8] J. H. McNally, J. W. Barnes, B. J. Dropesky, P. A. Seeger, and K. Wolfsberg, Phys. Rev. C (9), 717 (1974).

[9] W. Younes and H. C. Britt, Phys. Rev. C (67), 024610 (2003).

[10] J. B. Wilhelmy (2003), private communication.

[11] H. C. Britt, in *Proceedings of the Fourth International Symposium on the Physics and Chemistry of Fission*, Julich, West Germany (International Atomic Energy Agency, Vienna, 1979), vol. 1, p. 3.

[12] P. M. Endt and P. B. Smith, eds., *Nuclear Reactions* (North-Holland Publishing Company, Amsterdam, 1962), vol. 2, chap. 2.

TABLE I: Integral cross sections normalized to ^{235}U for the Tamped Flattop neutron flux (i.e., the ratio R_{235} described in the text). Our results, obtained from surrogate (t, pf) data are listed in column 4, and compared to the ENDL, ENDF/B-VI, and ENDF/B-VII evaluations in columns 5, 6, and 7, respectively.

Nucleus	J^π	E_x (keV)	Surrogate	ENDL	ENDF/B-VI	ENDF/B-VII
^{235}U	$7/2^-$	0.000	1.00			
	$9/2^-$	46.207	1.00			
	$11/2^-$	103.035	0.97			
^{235}U	$1/2^+$	0.077	0.83			
	$3/2^+$	13.040	0.88			
	$5/2^+$	51.709	0.85			
^{237}U	$1/2^+$	0.000	0.39	0.69	0.55	0.43
	$3/2^+$	11.39	0.42			
	$5/2^+$	56.30	0.40			
^{239}U	$5/2^+$	0.000	0.30	0.59		0.15
^{241}Pu	$5/2^+$	0.000	1.10	1.32	1.36	
^{243}Pu	$7/2^+$	0.000	0.91	0.45	0.45	

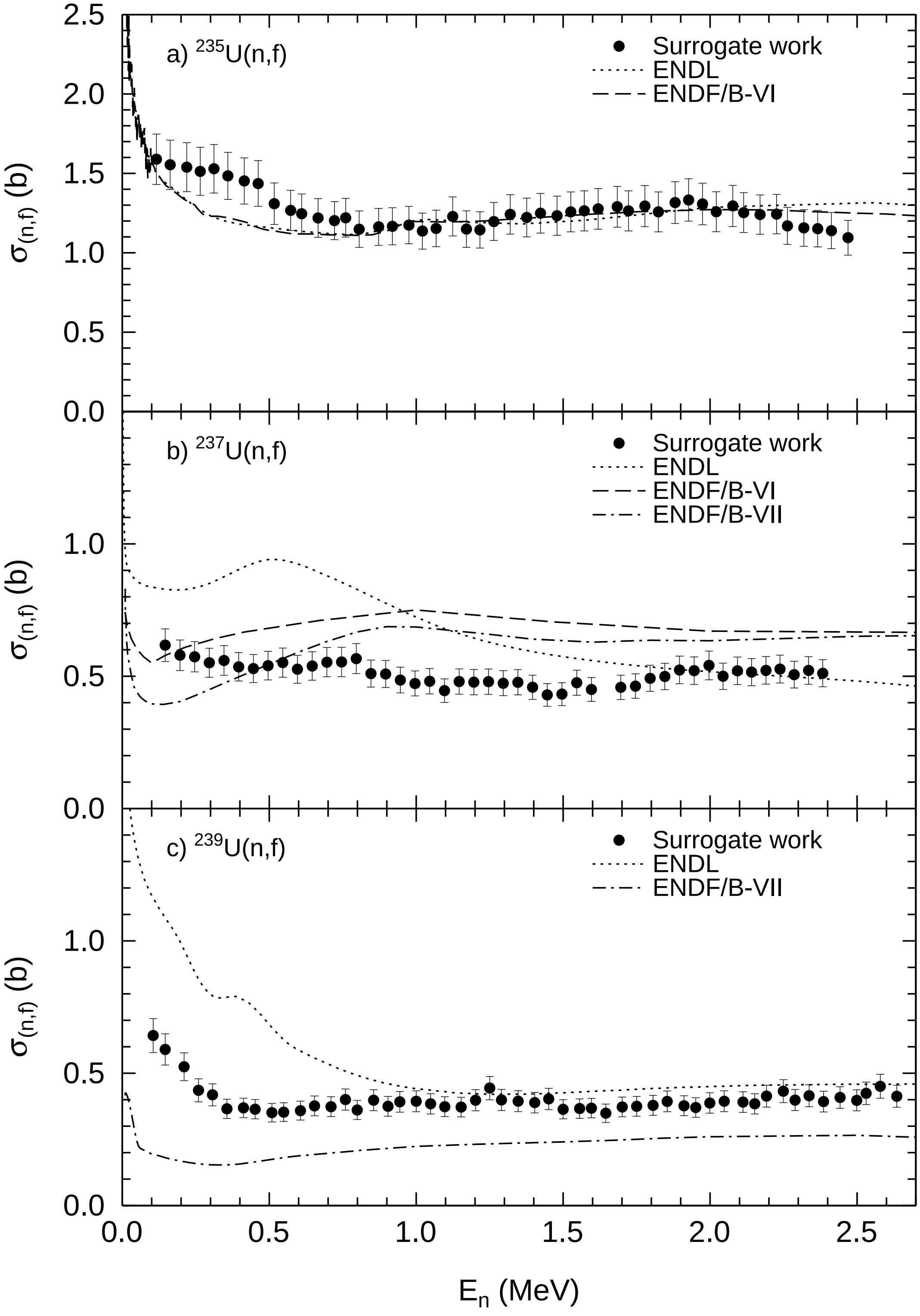

FIG. 1: Neutron-induced fission cross sections for targets of a) ^{235}U, b) ^{237}U, and c) ^{239}U, deduced [1, 2] from surrogate data, and compared to the ENDL [5], ENDF/B-VI [3], and ENDF/B-VII [4] evaluations.

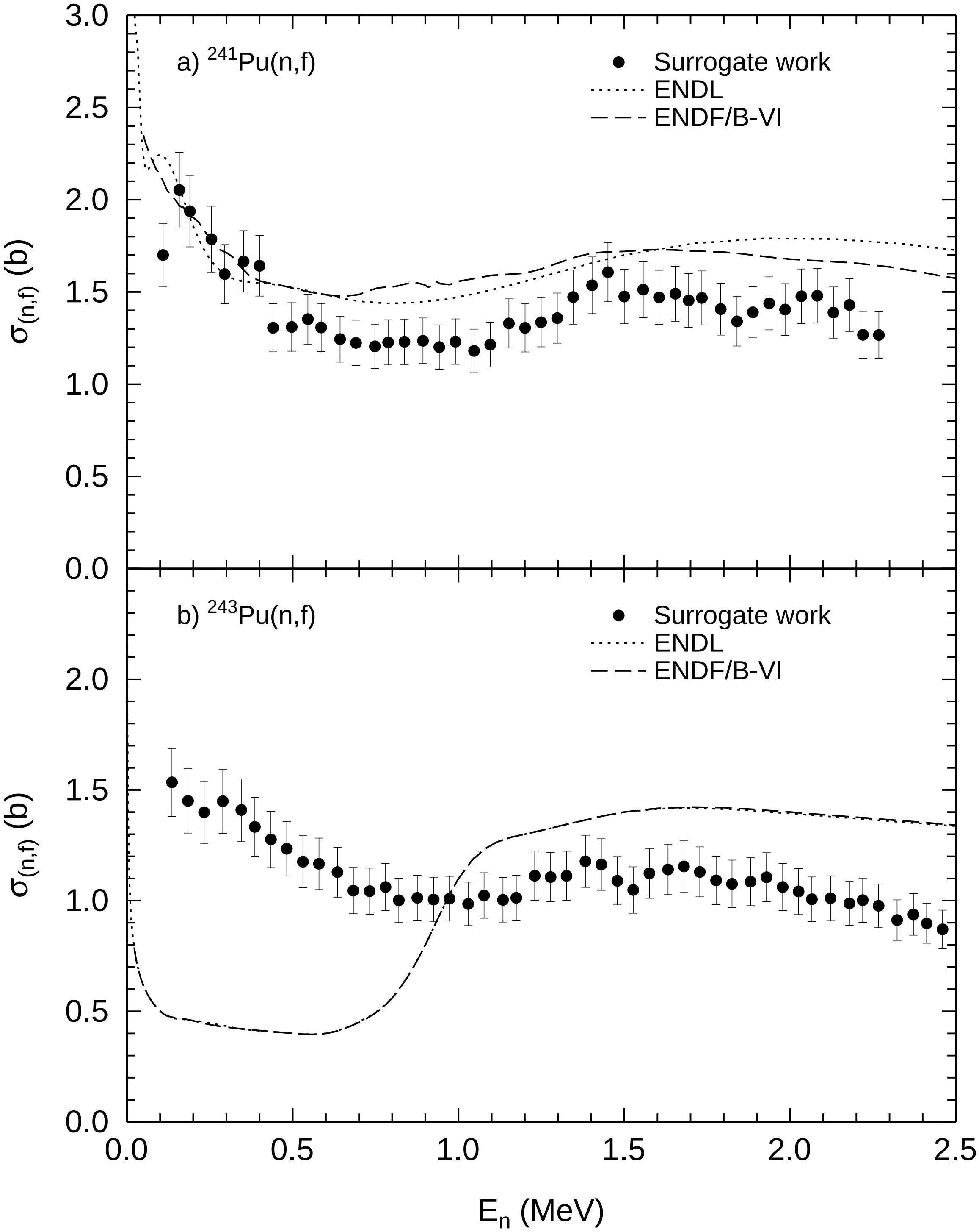

FIG. 2: Neutron-induced fission cross sections for targets of a) ^{241}Pu and b) ^{243}Pu, deduced [1, 2] from surrogate data, and compared to the ENDL [5] and ENDF/B-VI [3] evaluations. The ENDL and ENDF/B-VI curves in panel b) overlap.

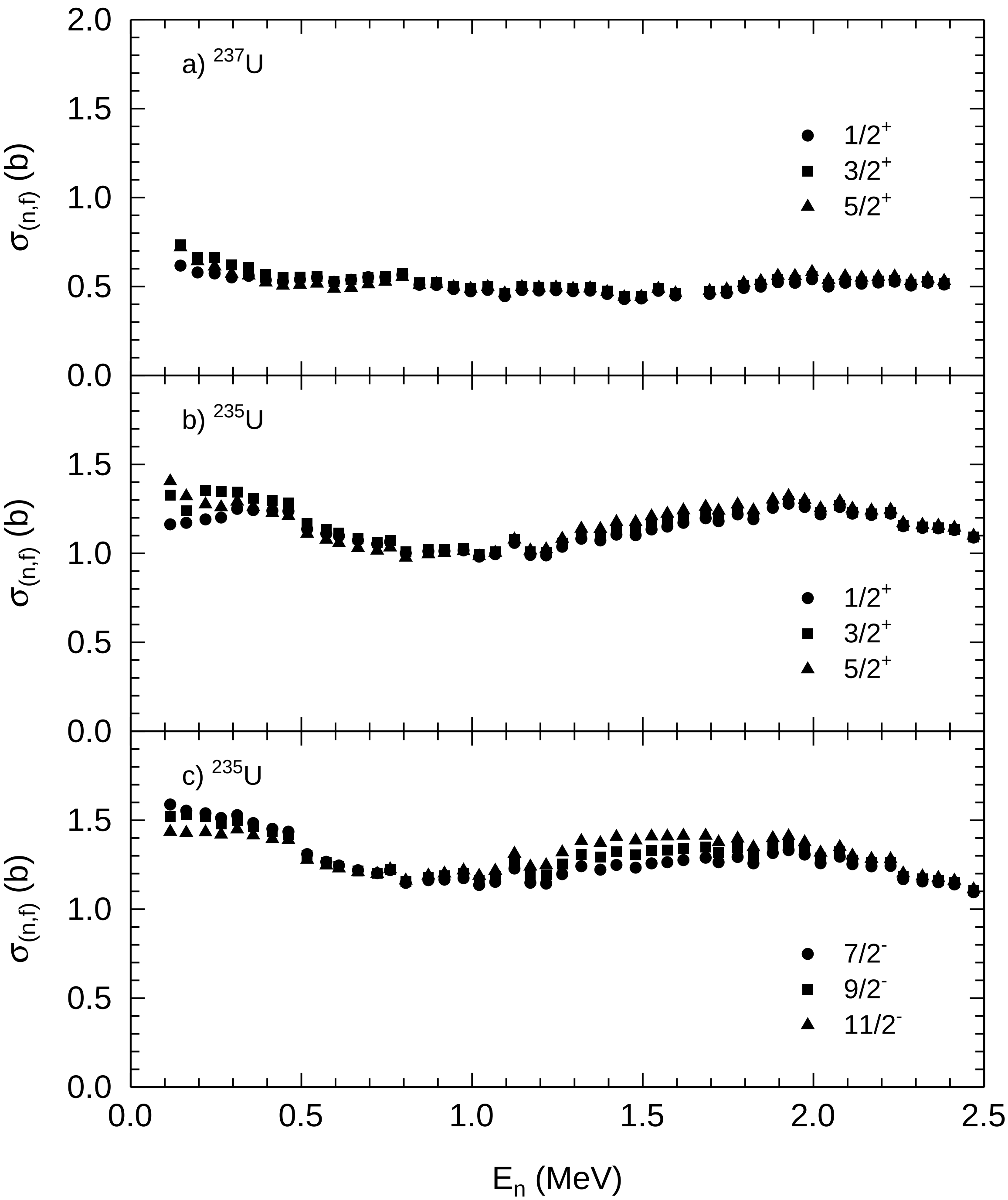

FIG. 3: Neutron-induced fission cross sections for excited states in a) the ground-state band of ^{237}U, b) the $T_{1/2} \approx$ 26-minute isomer-state band of ^{235}U, and c) the ground-state band of ^{235}U, deduced from surrogate (t, pf) data [1, 2].

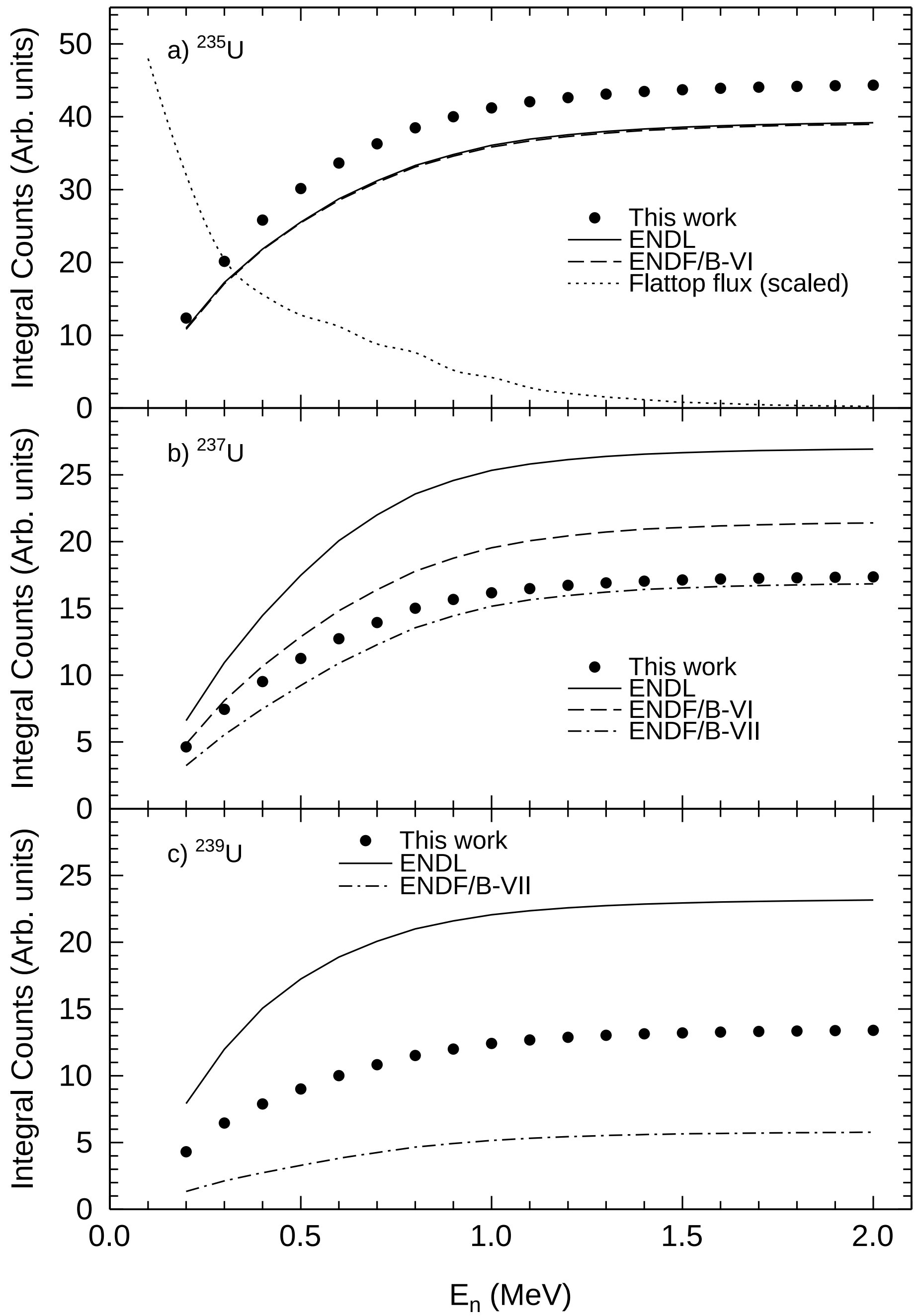

FIG. 4: Integrated cross sections for Neutron-induced fission, plotted as running sums, for targets of a) ^{235}U, b) ^{237}U, and ^{239}U, using a "Tamped Flattop" neutron flux folded with (n, f) cross sections deduced from our surrogate work [1, 2], and compared to the integrals obtained using (n, f) cross sections taken from ENDL [5], ENDF/B-VI [3], and ENDF/B-VII [4] evaluations. In panel a), the "Tamped Flattop" flux is scaled by an arbitrary factor and plotted. The ENDL and ENDF/B-VI curves in panel a) overlap.

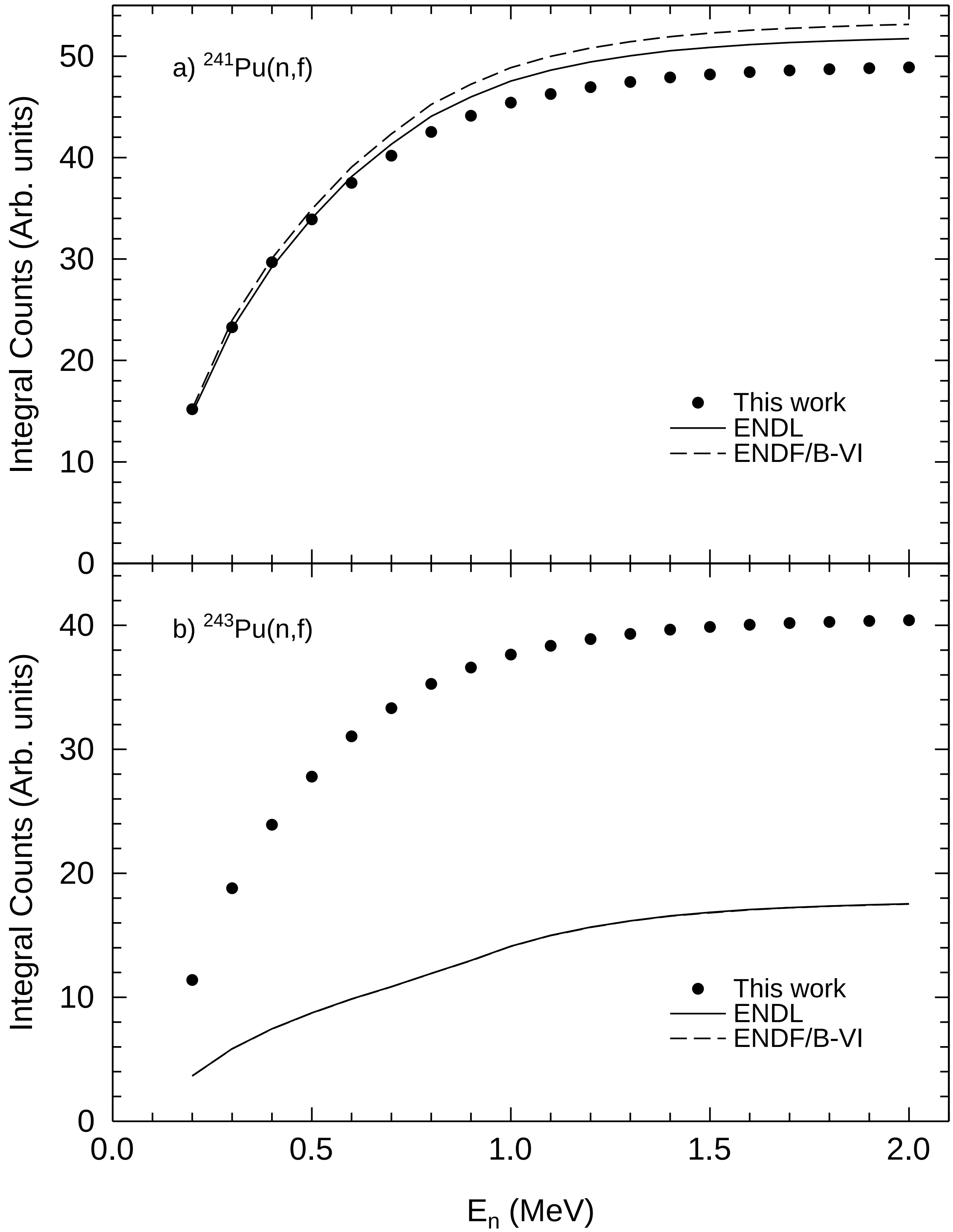

FIG. 5: Integrated cross sections for Neutron-induced fission, plotted as running sums, for targets of a) ^{241}Pu and b) ^{243}Pu, using a "Tamped Flattop" neutron flux folded with (n, f) cross sections deduced from our surrogate work [1, 2], and compared to the integrals obtained using (n, f) cross sections taken from ENDL [5] and ENDF/B-VI [3]. The ENDL and ENDF/B-VI curves in panel b) overlap.

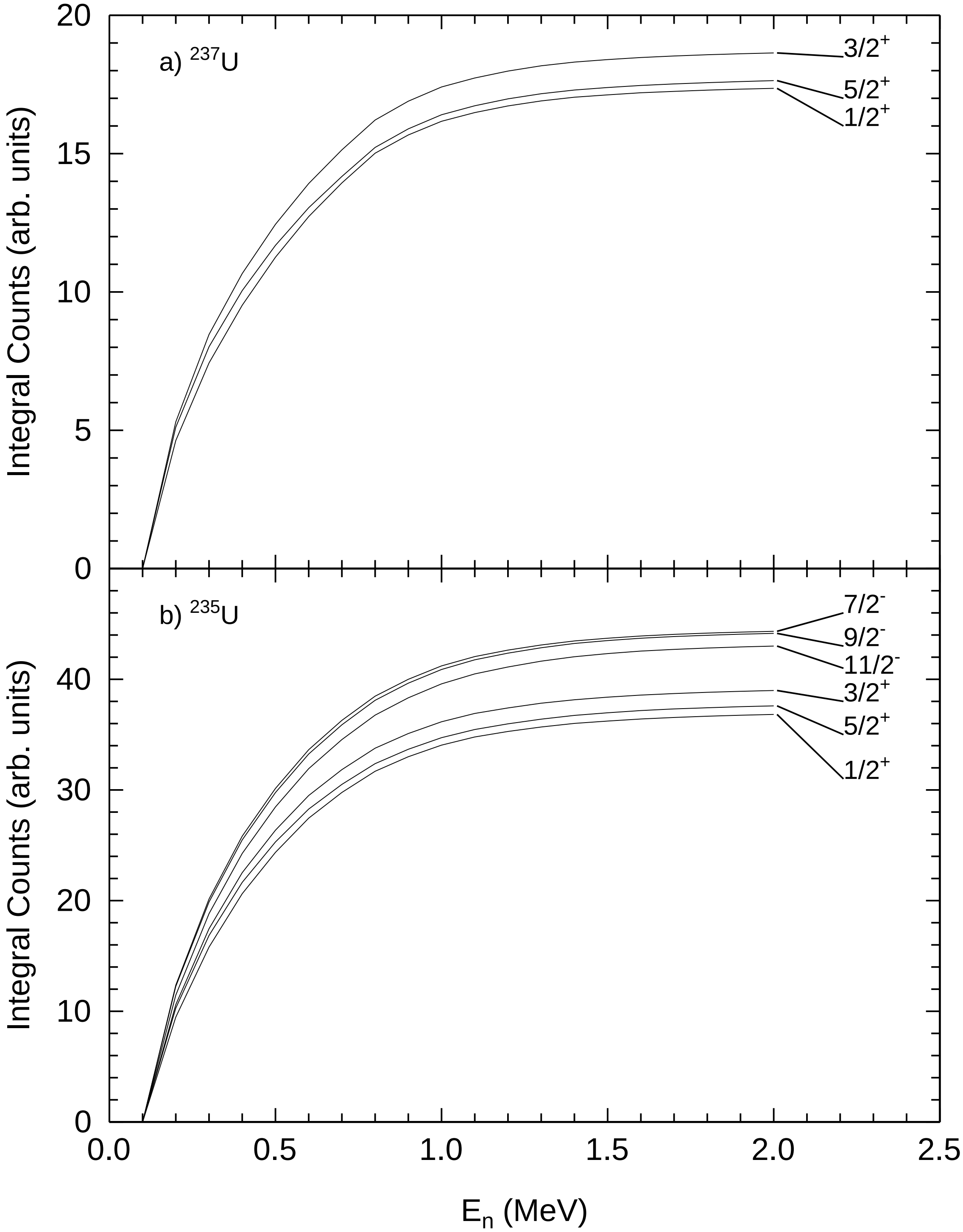

FIG. 6: Integrated cross sections for Neutron-induced fission, plotted as running sums, for excited states in a) the ground-state band of ^{237}U and b) the ground-state and isomer-state bands of ^{235}U, using a "Tamped Flattop" neutron flux folded with (n, f) cross sections deduced from our surrogate work [1, 2].

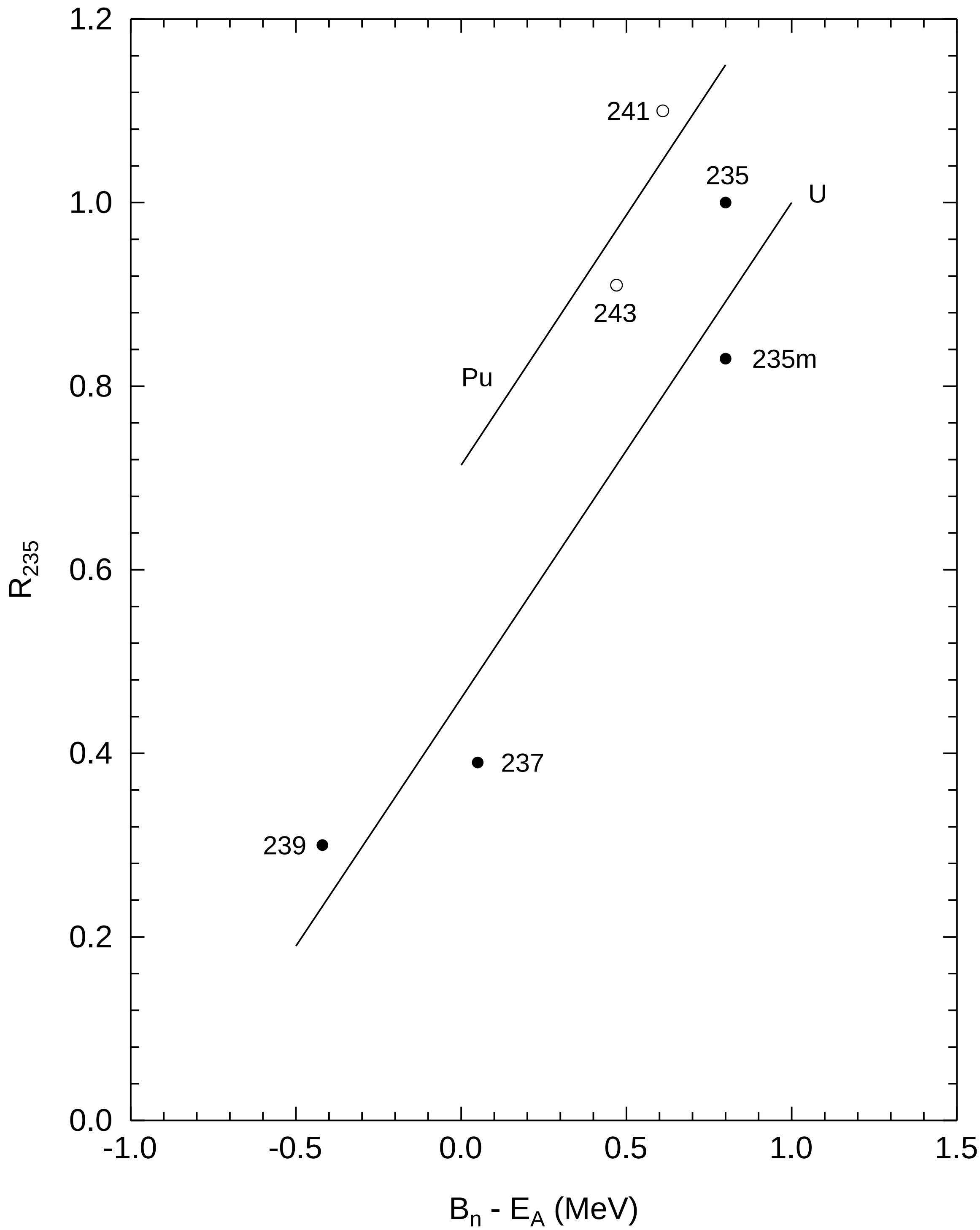

FIG. 7: Values of the ratio of integral cross sections R_{235} defined in the text for targets of Pu (open circles) and U (solid circles). The points are labeled by the mass number of the target appearing in the numerator of the R_{235} ratio. Parallel lines approximating the behavior of the R_{235} ratio as a function of $B_n - E_A$ have been drawn to guide the eye.